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The Steubenville Comprehensive Air Monitoring Program (SCAMP): Associations among Fine Particulate Matter, Co-Pollutants, and Meteorological Conditions

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ABSTRACT

We determined 24-hr average ambient concentrations of $PM_{2.5}$ and its ionic and carbonaceous components in Steubenville, OH, between May 2000 and May 2002. We also determined daily average gaseous co-pollutant concentrations, meteorological conditions, and pollen and mold spore counts. Data were analyzed graphically and by linear regression and time series models. Multiple-day episodes of elevated fine particulate matter ($PM_{2.5}$) concentrations often occurred during periods of locally high temperature (especially during summer), high pressure, or low wind speed (especially during winter) and generally ended with the passage of a frontal system. After removing autocorrelation, we observed statistically significant positive associations between concentrations of $PM_{2.5}$ and concentrations of CO, NO_x, and SO_2. Associations with NO_x and CO exhibited significant seasonal dependencies, with the strongest correlations during fall and winter. NO_x, CO, SO_2, O_3, temperature, relative humidity, and wind speed were all significant predictors of $PM_{2.5}$ concentration in a time-series model with external regressors, which successfully accounted for 79% of the variance in log-transformed daily $PM_{2.5}$ concentrations. Coefficient estimates for NO_x and temperature varied significantly by season. The results provide insight that may be useful in the development of future $PM_{2.5}$ reduction strategies for Steubenville. Additionally, they demonstrate the need for PM epidemiology studies in Steubenville (and elsewhere) to carefully consider the potential confounding effects of gaseous co-pollutants, such as CO and NO_x, and their seasonally dependent associations with $PM_{2.5}$.

IMPLICATIONS

The results presented here suggest that studies assessing the health effects of $PM_{2.5}$ in Steubenville, OH, must carefully consider the potential confounding effects of gaseous pollutants such as CO and NO_x, whose associations with $PM_{2.5}$ depend strongly on season. Several past epidemiological studies in Steubenville, which were instrumental in the promulgation of a new National Ambient Air Quality Standard (NAAQS) for $PM_{2.5}$, did not adequately consider these potential effects and must be interpreted cautiously. A new study should be conducted to better evaluate the effect of Steubenville's ambient air pollutant mixture on human health. The results might also be useful in the development of a $PM_{2.5}$ control strategy for Steubenville to meet the NAAQS.

INTRODUCTION

Fine particulate matter ($PM_{2.5}$), which consists of several diverse chemical constituents, coexists and potentially interacts with many other components in the air. Included among these are co-pollutant gases, such as CO, SO_2, NO_x, and O_3, which are regulated by the U.S. Environmental Protection Agency (EPA) because of the risks they pose to public health or the environment. These gases and other co-pollutants may be emitted by the same sources that emit $PM_{2.5}$ and its precursors, or themselves may serve as precursors to secondary $PM_{2.5}$ formation. Variations in this complex ambient mixture of pollutants are accompanied and potentially influenced by variations in an array of meteorological conditions. Hence, an analysis of associations among $PM_{2.5}$, co-pollutants, and meteorological conditions is prerequisite to understanding the formative mechanisms and effects of $PM_{2.5}$ at a particular location.

Several studies have identified factors that might contribute to ambient levels of $PM_{2.5}$. Anderson et al.[1] reported that high summertime concentrations of $PM_{2.5}$ at a site near Pittsburgh, PA, generally resulted from $PM_{2.5}$ transport to the site with the passage of frontal systems following periods of high atmospheric pressure. Chu and

Cox[2] found that episodes of elevated $PM_{2.5}$ concentrations in the eastern United States were generally associated with stagnant summertime high-pressure systems, and Vukovich and Sherwell[3] used meteorological data to identify possible differences in the processes affecting $PM_{2.5}$ mass at Washington, DC, and at a nearby remote site. Using a synoptic climatological approach, other studies[4,5] have assessed the impacts of weather on ambient levels of PM at various worldwide locations. Additionally, ambient measurements of particle component and gaseous co-pollutant concentrations have been used to study the processes leading to the secondary formation of these particle components.[6] Rizzo et al.[7] developed time-series transfer-function models describing PM_{10} concentrations as a function of O_3, NO_x, CO, and SO_2 concentrations for six U.S. cities to predict changes in ambient PM_{10} levels that would result from reductions in ambient O_3, which was used as a surrogate for photochemical activity. Studies such as these will be particularly important in the development of $PM_{2.5}$ reduction strategies for nonattainment areas to meet the National Ambient Air Quality Standard (NAAQS) for $PM_{2.5}$.[8]

Even more important are the implications that associations among $PM_{2.5}$, co-pollutants, and meteorological conditions might have for epidemiological studies that have associated $PM_{2.5}$ with adverse health effects. Such studies[9,10] served as a principal driving force for the promulgation of the NAAQS for $PM_{2.5}$. However, Moolgavkar and Luebeck[11] criticized several of these studies for failing to sufficiently consider the potential confounding effects of important co-pollutants and seasonality. Lipfert and Wyzga[12,13] also emphasized the need for epidemiological studies to consider co-pollutants, especially CO and NO_2, as potential causal factors and discussed problems that can arise from collinearity among pollutant variables even when appropriate co-pollutants are considered. In 1998, the National Research Council[14] stressed the need for additional research to better understand the interactions between PM and gaseous co-pollutants in relation to human health. Since then, several epidemiological studies[15-20] have considered the combined effects of co-pollutants and PM on public health and have discovered significant associations between mortality or morbidity and variables such as CO, NO_2, SO_2, O_3, temperature, and aeroallergens. In some instances, these associations were stronger than those between PM and the health endpoints being considered. In a meta-analysis synthesizing the results of 109 time-series studies of air pollution and mortality from around the world, Stieb et al.[21] concluded that "PM_{10}, CO, NO_2, O_3, and SO_2 were all positively and significantly associated with all-cause mortality." However, it is still unclear whether one of these pollutants, a combination of several pollutants, or some other factor is responsible for the observed health effects.

This paper examines associations among daily average concentrations of $PM_{2.5}$, gaseous co-pollutants (SO_2, CO, NO_x, O_3), and weather conditions that were measured at an outdoor ambient air monitoring site in Steubenville, OH, between May 2000 and May 2002 as part of the Steubenville Comprehensive Air Monitoring Program (SCAMP). $PM_{2.5}$ ionic and carbonaceous components, pollen, and mold spores are also considered. A previous paper[22] studied spatial and temporal variabilities and interrelationships among $PM_{2.5}$ and its ionic and carbonaceous components in the Steubenville region and found that the mean $PM_{2.5}$ concentration at Steubenville, based on 2 years of monitoring data, was 18.4 $\mu g/m^3$, or 3.4 $\mu g/m^3$ above the proposed annual $PM_{2.5}$ NAAQS of 15 $\mu g/m^3$. Hence, a $PM_{2.5}$ reduction strategy will likely be required for Steubenville. A study of the associations among $PM_{2.5}$, co-pollutants, and weather conditions is important for identifying factors that may contribute to the city's elevated $PM_{2.5}$ concentrations.

Furthermore, Steubenville was a focus of epidemiological studies that were important in establishing the foundation for the $PM_{2.5}$ NAAQS. For example, Schwartz and Dockery[23] reported that daily mortality was significantly associated with total suspended particulate (TSP) count but not with SO_2 in a multi-pollutant model using data collected at Steubenville, and Dockery et al.[9] reported an association between elevated ambient concentrations of fine particles at Steubenville and increased risk of mortality as part of the Harvard Six Cities Study. Reanalyses have called these results into question, however. Moolgavkar et al.[24] reanalyzed the Schwartz and Dockery data and found that inclusion of SO_2 in the model with TSP caused the effect of TSP on daily mortality to become statistically insignificant. The Health Effects Institute[25] reanalyzed the Six Cities Study data reported by Dockery et al.[9] and concluded that "The Six Cities Study, with its small number of cities and high degree of correlation among the air pollutants monitored, did not permit a clear distinction among the effects of gaseous and fine particle pollutants."[25]

Chock et al.[26] recently studied associations between air pollutant concentrations and daily mortality in Pittsburgh, PA, which is located ~58 km to the east of Steubenville. The analysis discovered important seasonal dependencies in correlation functions among air pollutant concentrations, illustrating the need to use seasonal models that consider the effects of both PM and gaseous co-pollutants. Moolgavkar et al.[24] also found season to be an important factor in their analysis of TSP, SO_2, and mortality at Steubenville. Moreover, when Chock et al.[26] used

such seasonal models, they uncovered significant multicollinearity problems among highly correlated concentrations of PM and co-pollutants, casting doubt on the results of nonseasonal models that indicated a significant association between PM_{10} and daily mortality.

In this paper, we use time-series analysis and regression techniques to study potential temporal dependencies in the associations among $PM_{2.5}$, co-pollutants, and weather conditions measured at Steubenville. On the basis of the findings of Chock et al.,[26] the results of this analysis might be useful in the design and interpretation of studies assessing the health effects of $PM_{2.5}$ in Steubenville.

METHODS

Experimental Methods

An overview of the SCAMP outdoor ambient air monitoring program was provided previously.[22] This paper focuses on 24-hr average $PM_{2.5}$, particulate ion, particulate carbon, gaseous co-pollutant, pollen, mold spore, and meteorological data collected between May 13, 2000, and May 14, 2002, at the central outdoor monitoring site in Steubenville, OH. The monitoring site was located on the campus of Franciscan University of Steubenville (40.362° N, 80.615° W) atop a bluff overlooking the Ohio River, in close proximity to several major industrial facilities. All daily data reported in this paper were collected and averaged from 9:00 a.m. on the reported day until 9:00 a.m. on the following day to be consistent with the indoor and personal sampling portion of SCAMP, which was conducted concurrently with the outdoor portion.

Ambient $PM_{2.5}$ concentrations were measured each day by the Federal Reference Method (FRM) for $PM_{2.5}$. Ambient concentrations of several ionic components (SO_4^{2-}, NO_3^-, and NH_4^+) of the $PM_{2.5}$ samples taken on every fourth day were determined by ion chromatography. In addition, fine particulate carbon species were collected every fourth day on a non-denuded quartz filter by use of a $PM_{2.5}$ speciation sampler, and ambient concentrations of elemental carbon (EC) and organic carbon (OC) were determined by a thermal optical transmittance procedure. All of these methods have been described previously in greater detail.[22] To approximately account for the mass of elements (such as H, O, and N) associated with carbon in organic compounds, we multiplied the concentration of OC by a factor of 1.4 to estimate the concentration of organic material (OM).

Ambient concentrations of gaseous priority pollutants, including SO_2, NO_x, CO, and O_3, were measured continuously by an Advanced Pollution Instruments (API) model 900 turnkey ambient air sampling system. The system consisted of individual rack-mounted gaseous pollutant analyzers, which were housed in a mobile trailer and integrated into a dedicated sampling manifold. Gaseous pollutants were collected through a typical glass "candy cane" at a sampling height of ~3.5 m above ground level. The gas analyzers were all API models and included an automated federal equivalent method (FEM) ultraviolet (UV) fluorescence SO_2 analyzer (model 100), an automated FRM chemiluminescence NO_x analyzer (model 200A), an automated FEM UV absorption O_3 analyzer (model 400), and an automated FRM gas filter correlation CO analyzer (model 300). The NO_x analyzer measured concentrations of NO_x and NO; concentrations of NO_2 were computed by difference. All gas measurements were made in accordance with quality assurance guidelines outlined by the EPA.[27,28] The performance of each analyzer was monitored by taking daily (shortly after midnight) calibration readings for zero and span concentrations of appropriate certified gas standards. Zero and span control charts were used to determine the validity of measured data and to determine when physical adjustments to the instruments were required. For each gaseous pollutant, hourly average concentrations recorded by the system's data logger were corrected to account for changes in instrument performance by linear interpolation between adjacent daily calibration readings. Daily average concentrations were computed as the arithmetic mean of these corrected hourly concentrations and were considered valid only if at least 19 valid hourly concentrations were available for the 24-hr sampling period.

Meteorological conditions, including wind speed, temperature, relative humidity (RH), solar radiation, and barometric pressure (BP), were monitored continuously by standard instrumentation (Met One) atop a 10-m tower. Again, daily average values for each of these parameters were computed as the arithmetic means of hourly averages recorded by the data logger and were considered valid only if at least 19 valid hourly values were available.

Pollen and mold spores were collected in a 7-day Burkard volumetric spore trap. Ambient air was sampled at a constant volumetric flow rate, and pollen and mold spores in the sampled stream impacted on an adhesive plastic tape, which rotated once at a constant rate during the course of each weekly sampling interval. Exposed tapes were analyzed by a certified environmental microbiology laboratory (Air Quality Sciences, Marietta, GA), which used optical microscopy counting procedures to determine speciated daily average pollen and mold spore counts.

Statistical Methods

Linear regression analysis was used to quantify the associations among $PM_{2.5}$, fine particle components, gaseous co-pollutants, and meteorological conditions. However,

Table 1. Sampling schedule and data recovery for 24-hr average data collected at the Steubenville monitoring site, as well as transformations and time-series models used to adjust these data.

Variable	Sampling Period (M/DD/YY)	Sampling Frequency ($days^{-1}$)	No. Valid Samples	Data Recovery (%)[a]	Trans.[b]	Mov. Ave.[c]	ARIMA Parameters[d] p	d	q
$PM_{2.5}$	5/13/00–5/14/02	1/1	640	87	$\ln(x)$	–	1	1	1,2
SO_4^{2-}	5/16/00–5/14/02	1/4	151	83	$\ln(x)$	21	–	–	–
NO_3^-	5/16/00–5/14/02	1/4	151	83	$\ln(x)$	–	0	1	1
NH_4^+	5/16/00–5/14/02	1/4	151	83	$\ln(x)$	–	–	–	–
EC	8/12/00–5/14/02	1/4	146	91	$x^{0.25}$	–	–	–	–
OM	8/12/00–5/14/02	1/4	146	91	$x^{0.25}$	–	–	–	–
SO_2	5/16/00–5/14/02	1/1	624	86	$\ln(x)$	–	0	0	1
NO	5/16/00–5/14/02	1/1	569	78	$\ln(x)$	–	1,2,3	0	1
NO_2[e]	5/16/00–5/14/02	1/1	504	69	$x^{0.5}$	–	0	0	1,6
NO_x	5/16/00–5/14/02	1/1	525	72	$\ln(x)$	–	1,2,3	0	1
CO	5/16/00–5/14/02	1/1	620	85	$\ln(x + 0.1)$	–	0	1	1,2
O_3	5/16/00–5/14/02	1/1	580	80	$x^{0.5}$	–	0	1	1,2
WS	5/16/00–5/14/02	1/1	697	96	$\ln(x)$	–	1,2,3	0	1
Temp	5/16/00–5/14/02	1/1	697	96	–	–	1	1	1,2
RH	5/16/00–5/14/02	1/1	696	95	$x^{1.5}$	–	0	1	1,2,3
Rad	5/16/00–5/14/02	1/1	605	83	–	–	1,2,3	1	1
BP	5/16/00–5/14/02	1/1	697	96	–	–	1,10	1	1,2
Pollen[f]	6/21/00–5/13/02	1/1	586	85	NA	NA	NA	NA	NA
Mold spores[f]	6/21/00–5/13/02	1/1	599	87	NA	NA	NA	NA	NA

Notes: [a]Actual number of valid samples/potential number of valid samples based on sampling period and frequency; [b]Data transformation; [c]Indicates that a moving average of the specified number of measurements was subtracted from the transformed data to stationarize the series; [d]p and q indicate the individual lags (see sampling frequency for units) to which autoregressive and moving average parameters, respectively, were fit; d indicates the order of differencing used in the ARIMA model; [e]Computed as $NO_x - NO$; [f]These data were not transformed or modeled; WS = wind speed; Temp = temperature; Rad = solar radiation; NA = not applicable.

as discussed by Connell et al.,[22] time series of data often must be transformed and modeled to remove autocorrelation before regression analysis is applied in order to better satisfy the assumption that the error component of the regression model is normally distributed, homoskedastic, and statistically independent.

Therefore, before performing linear regression analysis, we transformed variables as necessary to render their distributions more normal and to improve homoskedasticity. For each variable, we used time series plots, autocorrelation function (ACF) plots, and partial ACF plots in the standard way to identify serial dependencies. Autocorrelation at early lags was modeled using an autoregressive integrated moving average (ARIMA) model.[29] When estimating model parameters, we used a state-space representation of the process to handle missing values.[30] Nonstationarities, including seasonal patterns, were removed by the inclusion of first-order differencing in the ARIMA model or by subtracting out an approximately 3-month moving average before modeling, as needed. A suitable model was selected by minimizing the Aikake Information Criterion, provided that all estimated parameters were reasonable and statistically significant. Throughout this paper, statistical significance was determined at a significance level (α) of 0.05. An ACF plot and Ljung–Box tests for randomness were used to verify the statistical independence of residuals from the model. The residuals obtained by transforming and modeling (where necessary) a variable according to this procedure are hereafter referred to as the "adjusted" values of that variable. These adjusted values were used in the linear regression analyses. Therefore, the regression analyses examine associations among day-to-day fluctuations in the magnitudes of the variables being considered, rather than correlations that may be induced by similar or dissimilar seasonal cycles or temporal self-dependencies among these variables.

The transformation and the form of the ARIMA model used to adjust each variable are shown in Table 1. Figure 1 demonstrates the improvement afforded by the use of adjusted data in the linear regression analysis of $PM_{2.5}$ as a function of NO_x. The ACF plot of residuals from regression analysis using raw data (Figure 1a) shows that these residuals are highly autocorrelated and violate the assumption of statistical independence. When adjusted data are used (Figure 1b), the assumption of statistical

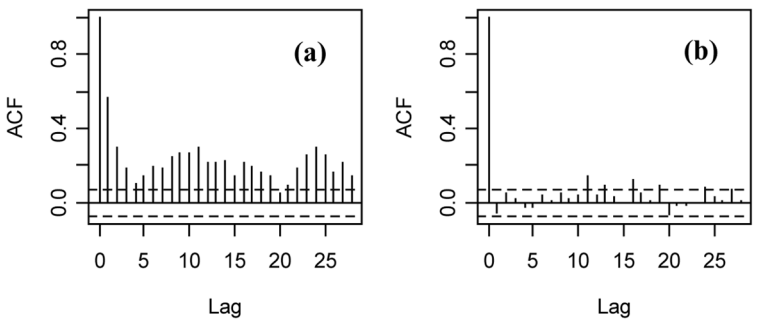

Figure 1. ACF plots of residuals from simple linear regression of $PM_{2.5}$ as a function of NO_x using raw data (a) and adjusted data (b). Lags are in units of days. Lines extending beyond the dashed interval indicate statistically significant autocorrelations.

independence is much better satisfied. In several instances, despite the use of adjusted data, regression model residuals exhibited a small amount of autocorrelation at an early lag. We reanalyzed a few of these instances by constructing a single ARIMA model with an external regressor, and the slope and intercept estimates and standard errors obtained by this approach did not differ significantly from those obtained by simple linear regression with adjusted data. Hence, these simple linear regression results are presented in this paper. A single ARIMA model with external regressors is presented, however, to examine the combined association between multiple gaseous co-pollutant and meteorological variables and $PM_{2.5}$ at Steubenville.

RESULTS AND DISCUSSION

Descriptive statistics for all valid 24-hr average $PM_{2.5}$ concentrations, gaseous co-pollutant concentrations, and weather conditions measured at the Steubenville monitoring site between May 2000 and May 2002 are presented in Table 2. Descriptive statistics are also provided for the subset of data collected on the 64 days with $PM_{2.5}$ concentrations above the 90th percentile value (34.7 $\mu g/m^3$). Negative values were obtained for several daily average CO concentration measurements that otherwise met all validation criteria, indicating that ambient CO concentrations were smaller in magnitude than instrument noise on these days. Although negative concentrations clearly do not represent physical reality, these negative values were used in the analyses presented in this paper because manipulating them would introduce bias.

Ambient concentrations of criteria air pollutant gases were generally well below regulatory levels set by the EPA. For example, no daily average NO_2 concentration exceeded the annual NO_2 NAAQS of 53 ppb, and the overall mean and maximum 24-hr average SO_2 concentrations were well below the annual and 24-hr SO_2 NAAQS of 30 and 140 ppb, respectively. Nonetheless, as shown in Table 2, days with the highest $PM_{2.5}$ concentrations at Steubenville during SCAMP often also had above-average concentrations of SO_2, NO_x, and CO. The mean and median daily concentrations of each of these gases computed for the 64 highest $PM_{2.5}$ days were greater than the 75th percentile concentrations computed with all data, and the 25th percentile concentrations computed for these 64 days were greater than the overall 50th percentile concentrations. This apparent positive association among constituents of the ambient air pollution mixture has important implications for understanding the formative mechanisms and possible health effects of $PM_{2.5}$ and will be analyzed further in this paper.

Table 2. Descriptive statistics for 24-hr average $PM_{2.5}$, gaseous co-pollutant, and weather data measured at the Steubenville monitoring site.

		All Data								10% Highest $PM_{2.5}$ Days				
Variable	**Units**	***n***	**Mean**	**SD**	**Min**	**Q25[a]**	**Q50**	**Q75**	**Max**	***n***	**Mean**	**Q25**	**Q50**	**Q75**
$PM_{2.5}$	$\mu g/m^3$	640	18.4	11.2	2.7	10.1	15.3	23.3	64.8	64	43.3	37.9	41.3	48.0
SO_2	ppb	624	10.5	8.4	1.3	5.0	8.2	12.9	60.1	47	22.7	11.1	19.5	31.3
NO	ppb	569	6.8	9.3	0.1	1.3	3.3	8.1	71.5	44	19.8	4.2	10.6	34.8
NO_2	ppb	504	12.7	5.8	1.9	8.3	11.8	16.4	33.1	32	18.4	15.5	18.7	22.3
NO_x	ppb	525	18.8	12.8	2.4	9.9	15.3	23.4	102.6	34	34.1	19.0	28.4	46.4
CO	ppm	620	0.30	0.35	−0.08	0.10	0.20	0.36	2.73	52	0.84	0.36	0.61	1.22
O_3	ppb	580	28.0	12.6	2.4	19.3	26.6	35.5	77.0	54	28.8	13.7	27.0	41.4
WS	m/sec	697	2.52	1.06	0.78	1.71	2.33	3.16	7.68	62	2.01	1.31	1.94	2.55
Temp	°C	697	11.0	9.5	−14.9	3.3	12.0	19.0	28.1	62	17.4	13.4	16.6	24.0
RH	%	696	71.95	12.09	32.45	64.23	72.96	79.98	99.57	62	72.83	65.77	75.06	78.63
Rad	W/m^2	605	152	94	1	73	138	229	367	44	190	116	177	270
BP[b]	mmHg	697	733.3	4.3	719.1	730.8	733.6	736.3	744.2	62	735.9	734.1	735.8	737.6

Notes: Statistics were calculated with all valid data collected during SCAMP, as well as with only data collected during the 64 days for which $PM_{2.5}$ concentrations at Steubenville were above the 90th percentile concentration; [a]Q25, Q50, and Q75 indicate the 25th, 50th, and 75th quartiles; [b]Not corrected to sea level; measurements were taken ∼315 m above sea level; WS = wind speed; Temp = temperature; Rad = solar radiation.

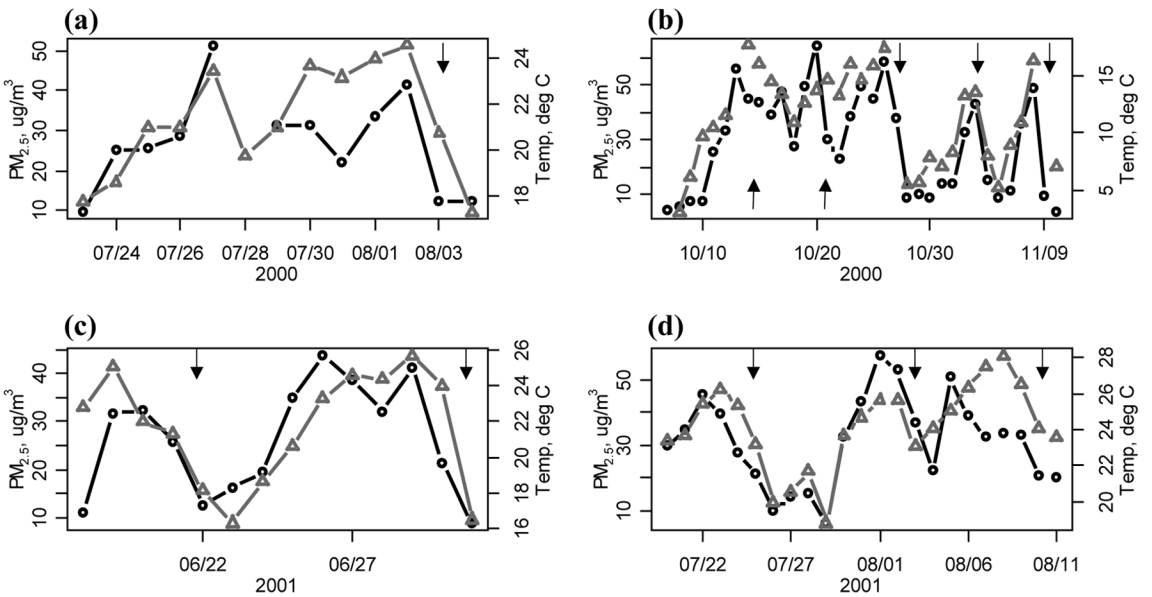

Figure 2. Time series of 24-hr average $PM_{2.5}$ concentrations (circles) and temperatures (triangles) measured at the Steubenville monitoring station during four periods: July 23 through August 4, 2000 (a); October 7 through November 10, 2000 (b); June 18 through July 1, 2001 (c); and July 20 through August 11, 2001 (d). Arrows indicate days on which cold fronts passed through the area.

In addition to the data presented in Table 2, speciated ambient pollen and mold spore counts (number/m^3) were determined at the Steubenville monitoring site because these aeroallergens may be important contributors to respiratory health effects. Pollen and spores have been identified as components of coarse PM,[31,32] and evidence of microspores and pollen- and spore-derived cytoplasmic material has been found in fine PM.[32] Nevertheless, the contribution of pollen and mold spores to PM mass could not be quantified in this study because of the absence of size-fractionated pollen and mold spore mass data. Instead, daily average counts of total pollen, total mold spores, and individual pollen and mold spore species were compared with daily average $PM_{2.5}$ concentrations by use of descriptive statistics and graphs. No obvious associations were discovered, but these data might be useful for incorporation in future epidemiological studies that attempt to better account for the effects of the many diverse constituents of the ambient air pollution mixture.

Episodes of Elevated $PM_{2.5}$ Concentration

Before performing regression analysis to examine associations between $PM_{2.5}$ and gaseous co-pollutants and weather variables, we examined several episodes of elevated $PM_{2.5}$ concentrations in depth to pinpoint conditions that accompanied and potentially contributed to these episodes. Episodes were defined as multiple-day events having at least one daily $PM_{2.5}$ concentration above the 95th percentile concentration (41.3 $\mu g/m^3$) measured during SCAMP and at least one above the 75th percentile concentration (23.3 $\mu g/m^3$). For each episode, daily $PM_{2.5}$ concentrations were compared with daily gas concentrations and weather conditions measured at the Steubenville monitoring site by use of time-series plots. Additionally, archived meteorological surface maps[33] were examined to determine the locations of frontal systems and centers of high and low pressure.

Several episodes exhibited a positive association between $PM_{2.5}$ concentration and temperature. This behavior was generally observed during the warmer portions of the year. Four such episodes, which occurred in July and August 2000, October and November 2000, June and July 2001, and July and August 2001, are shown in Figure 2. During each of these episodes, daily average temperatures at Steubenville varied by at least 7 °C and daily average $PM_{2.5}$ concentrations varied by at least 35 $\mu g/m^3$. Although the correlations between $PM_{2.5}$ concentration and temperature were not perfect and were at times slightly lagged, periods of locally elevated temperatures generally tended to correspond to periods of elevated $PM_{2.5}$ concentrations. Further insight is gained by considering days on which cold fronts passed through the Steubenville region. These days are denoted with arrows in Figure 2. As shown in Figure 2, $PM_{2.5}$ concentrations and temperatures both tended to increase between occurrences of these frontal systems, often peaking just before the passage of a front. Values of both variables decreased, sometimes sharply, with the frontal passage and often remained relatively low for 1 or more days thereafter.

During several other $PM_{2.5}$ episodes, we observed remarkable correlations between $PM_{2.5}$ concentrations and concentrations of CO, NO_x, and SO_2. These correlations

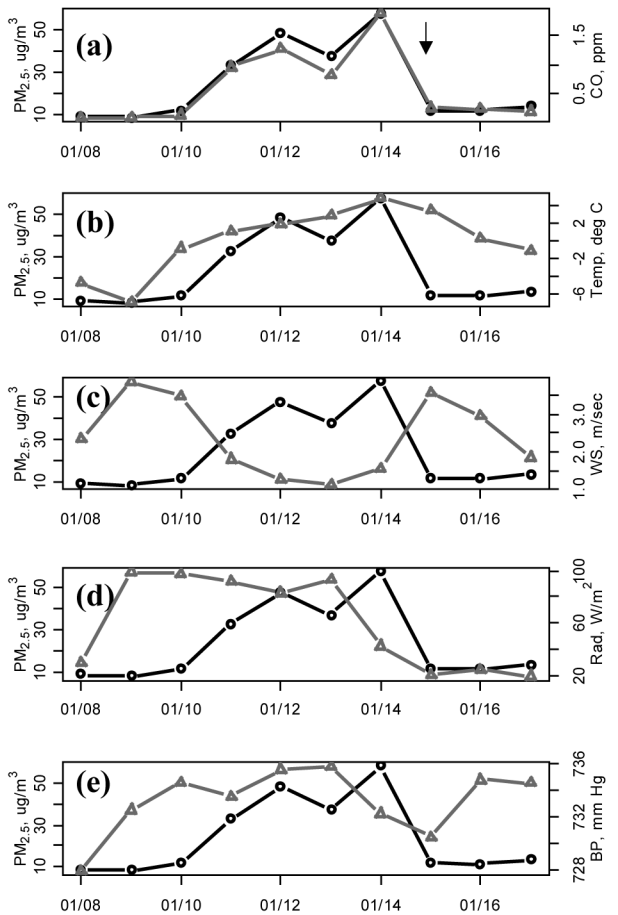

Figure 3. Time series of 24-hr average $PM_{2.5}$ concentrations (circles) and CO concentrations (a), temperatures (b), wind speed (WS; c), solar radiation (Rad; d), and BP (e), all indicated by triangles, measured at the Steubenville monitoring station between January 8 and January 17, 2001. Arrow in panel a denotes the passage of a frontal system through the area.

were most frequently observed during the cooler portions of the year. Three representative examples are presented in Figures 3, 4, and 5. During each of these $PM_{2.5}$ episodes, concentrations of the gases that are shown exceeded their respective overall 95th percentile values (0.99 ppm for CO, 42.8 ppb for NO_x, and 28.1 ppb for SO_2) at least once. Valid data were not available for NO_x and SO_2 between January 8 and January 17, 2001; however, as shown in Figure 3a, daily fluctuations in ambient CO concentrations corresponded very closely to daily fluctuations in $PM_{2.5}$ concentrations during this period. Figures 5a–5c, which present data from a period of January 2002, illustrate a strong association among day-to-day levels of $PM_{2.5}$, SO_2, NO_x, and CO during an apparent ambient air pollution episode. Figures 4a–4c, which show data from November 2001, also indicate simultaneous occurrences of local maxima in the ambient concentrations of these four pollutants.

Meteorological conditions were studied for these episodes to permit better understanding of the ambient conditions associated with each. As shown in Figures 3a and 3b, abnormally elevated $PM_{2.5}$ and CO concentrations occurred during an early January period when daily average temperatures reached a local maximum above 0 °C. Winds were relatively calm during the 4-day period having the highest ambient pollutant concentrations, but were considerably stronger immediately before and after this period (Figure 3c). The BP was high and skies were clear when concentrations of $PM_{2.5}$ and CO began to increase (Figures 3d and 3e), but BP and solar radiation began to decrease on January 14, when concentrations of both of these pollutants reached their highest peaks. Surface maps confirm that these decreases in BP and solar radiation accompanied the approach of an occluded front, which passed through Steubenville between 7:00 a.m. and 7:00 p.m. on January 15. Ambient concentrations of both $PM_{2.5}$ and CO fell sharply on this date.

Weather conditions during the November 2001 and January 2002 episodes presented in Figures 4 and 5, respectively, exhibited some similar characteristics. As

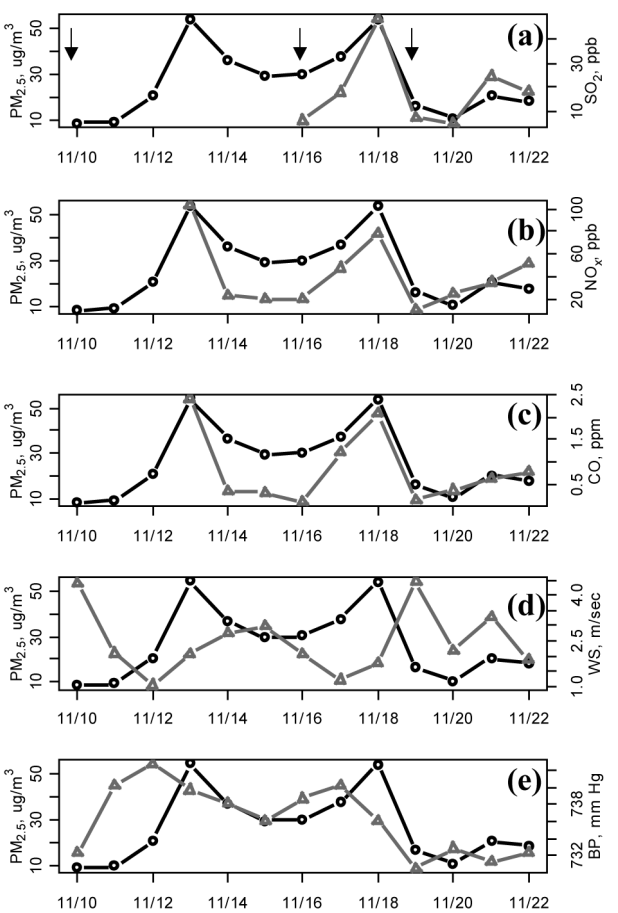

Figure 4. Time series of 24-hr average $PM_{2.5}$ concentrations (circles) and SO_2 concentrations (a), NO_x concentrations (b), CO concentrations (c), wind speed (WS; d), and BP (e), all indicated by triangles, measured at the Steubenville monitoring station between November 10 and November 22, 2001. Arrows in panel a denote the passage of frontal systems through the area.

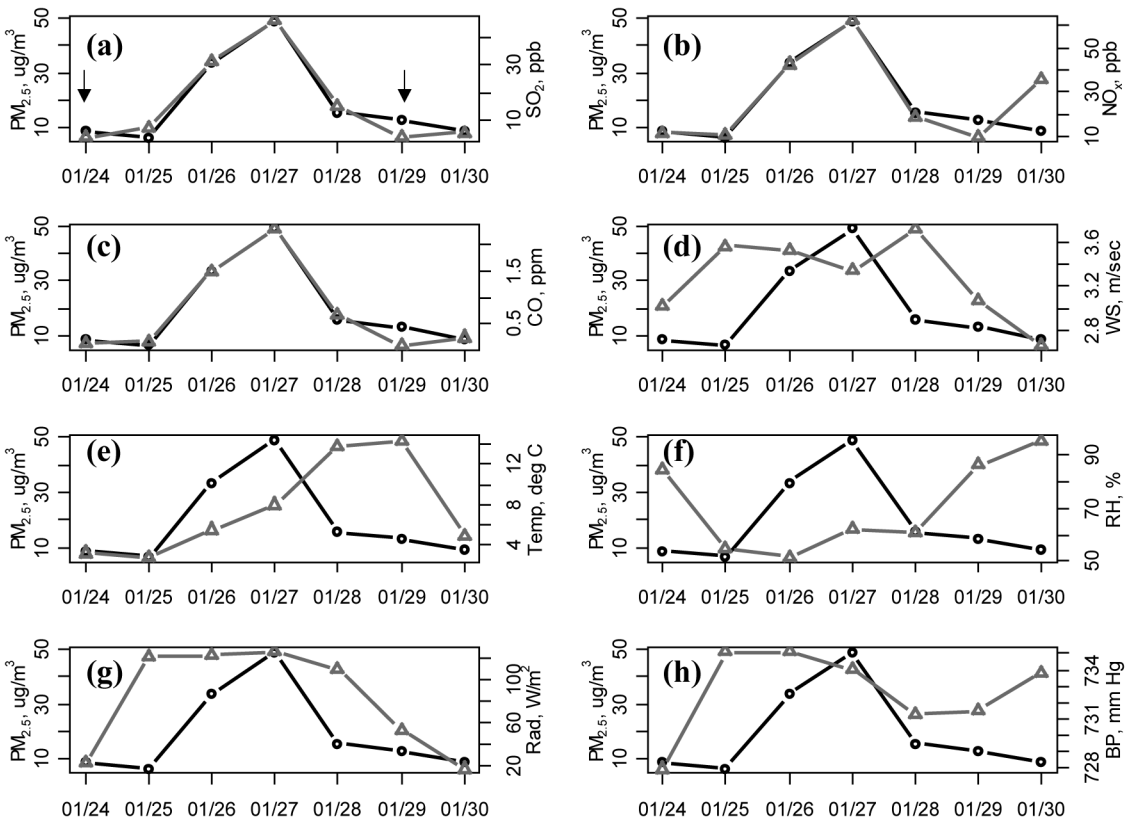

Figure 5. Time series of 24-hr average $PM_{2.5}$ concentrations (circles) and SO_2 concentrations (a), NO_x concentrations (b), CO concentrations (c), wind speed (WS; d), temperatures (e), RH (f), solar radiation (Rad; g), and BP (h), all indicated by triangles, measured at the Steubenville monitoring station between January 24 and January 30, 2002. Arrows in panel a denote the passage of frontal systems through the area.

shown in Figures 4e and 5h, during both of these episodes, $PM_{2.5}$ and co-pollutant concentrations increased during periods of high pressure and peaked just as BP began to decrease with the approach of a frontal system. Rapid decreases in ambient concentrations of $PM_{2.5}$, CO, NO_x, and SO_2 coincided with the passage of a cold front on November 19, 2001 (Figure 4) and the approach of a front on January 28 and its passage on January 29, 2002 (Figure 5). Additionally, Figure 4d is consistent with Figure 3c in showing a negative association between $PM_{2.5}$ concentration and surface wind speed. Like Figure 3, Figure 5 shows a January $PM_{2.5}$ episode that occurred when skies were relatively clear (Figure 5g) and daily average temperatures were above 0 °C (Figure 5e), although the local maximum temperature lags the local maximum $PM_{2.5}$ concentration in Figure 5e.

These findings provide insight into factors that might influence ambient levels of $PM_{2.5}$ in Steubenville. Studies of the episodes presented in Figures 2 through 5 all suggest that elevated $PM_{2.5}$ concentrations tended to occur during periods when no frontal systems passed through the region, with the end of an episode often coinciding with the passage of a front. BP frequently exhibited a unique lagged relationship with $PM_{2.5}$ as $PM_{2.5}$ concentrations tended to increase during a period of locally high pressure, remain elevated as pressure dropped with the approach of a front, and then drop as pressure increased after the passage of the front. These observations are likely indicative of the influence of surface high-pressure systems and associated aloft ridges on $PM_{2.5}$ concentrations in Steubenville. Subsidence, stagnation, and recirculation associated with these systems can yield poor ventilation conditions, allowing $PM_{2.5}$ concentrations to build up, and clear skies characteristic of high-pressure regimes may promote increased photochemical production of secondary $PM_{2.5}$. Decreases in $PM_{2.5}$ concentrations corresponding to the passage of frontal systems are likely attributable to the arrival of less-polluted air masses or to enhanced vertical mixing and surface winds associated with the frontal systems that improve the vertical and horizontal dispersion of pollutants.

The case studies above also suggest that the highest $PM_{2.5}$ concentrations often occurred during periods of locally elevated temperatures, particularly in warmer portions of the year, and during periods of relatively calm winds, particularly in cooler portions of the year. Jung et al.[34] similarly observed that elevated $PM_{2.5}$ concentrations in central and southeastern Ohio tended to occur during periods of high temperature and low wind speed. As with associations among $PM_{2.5}$, temperature, and wind speed, the occurrence of strong episodic associations among concentrations of

Table 3. Simple linear regression results for adjusted $PM_{2.5}$ as a function of adjusted gaseous co-pollutants or meteorological conditions.

Independent Variable	Overall	Winter	Spring	Summer	Fall
SO_2					
R^2	0.35	0.35	0.36	0.24	0.48
Slope	0.40	0.38	0.40	0.34	0.49
p	**<0.0001**[a]	**<0.0001**[a]	0.8240[b]	0.5844[b]	0.0763[b]
NO					
R^2	0.25	0.53	0.24	0.0014	0.53
Slope	0.22	0.28	0.19	−0.015	0.39
p	**<0.0001**[a]	**<0.0001**[a]	**0.0371**[b]	**0.0014**[b]	**0.0129**[b]
NO_2					
R^2	0.34	0.51	0.27	0.086	0.61
Slope	0.37	0.42	0.29	0.20	0.57
p	**<0.0001**[a]	**<0.0001**[a]	**0.0480**[b]	**0.0012**[b]	**0.0164**[b]
CO					
R^2	0.46	0.60	0.45	0.24	0.60
Slope	0.51	0.66	0.56	0.27	0.61
p	**<0.0001**[a]	**<0.0001**[a]	0.1701[b]	**<0.0001**[b]	0.4204[b]
O_3					
R^2	0.029	0.25	0.017	0.12	0.13
Slope	−0.090	−0.29	−0.067	0.17	−0.21
p	**0.0001**[a]	**<0.0001**[a]	**0.0009**[b]	**<0.0001**[b]	0.1762[b]
WS					
R^2	0.14	0.23	0.12	0.0094	0.20
Slope	−0.50	−0.67	−0.45	−0.12	−0.62
p	**<0.0001**[a]	**<0.0001**[a]	0.1455[b]	**0.0005**[b]	0.7574[b]
Temperature					
R^2	0.17	0.16	0.16	0.48	0.12
Slope	0.052	0.041	0.045	0.14	0.052
p	**<0.0001**[a]	**<0.0001**[a]	0.7296[b]	**<0.0001**[b]	0.3373[b]
RH					
R^2	0.0008	0.019	0.0000	0.0003	0.0000
Slope	-9.6×10^{-5}	-4.8×10^{-4}	1.3×10^{-5}	-7.9×10^{-5}	7.8×10^{-6}
p	0.4998[a]	0.0909[a]	0.1903[b]	0.4492[b]	0.2091[b]
Rad					
R^2	0.049	0.071	0.011	0.077	0.16
Slope	1.9	3.0	0.64	2.0	5.9
p	**<0.0001**[a]	**0.0007**[a]	**0.0208**[b]	0.4143[b]	**0.0367**[b]
BP					
R^2	0.012	0.018	0.0055	0.0013	0.024
Slope	0.42	0.43	0.28	−0.21	0.63
p	**0.0073**[a]	0.0954[a]	0.7205[b]	0.3043[b]	0.5794[b]

Notes: For each independent variable, regression was performed first with all data collected during SCAMP and then with subsets of these data collected during each of the four major seasons; Winter was used as the base case for comparing slope by season; Statistically significant *p* values are shown in bold; Intercept values were estimated, but are not shown; [a]*p* value resulting from a test of the hypothesis that the slope of the regression line is equal to zero; [b]*p* value resulting from a test of the hypothesis that the slope of the regression line for the season under consideration is equal to the slope of the regression line for winter; WS = wind speed; Rad = solar radiation.

$PM_{2.5}$ and primarily emitted pollutant gases (i.e., CO, NO_x, and SO_2) appeared to depend on season: most occurrences were observed during climatologically cool portions of the year. It is possible that these associations are the result of a common emission source of all four pollutants; however, major emissions of SO_2 and CO are not generally expected to originate from a common source. Given the meteorological observations presented above, a more plausible hypothesis is that reduced mixing heights, possibly because of surface inversions occurring on long, clear, calm nights, drive cool-season episodes of high $PM_{2.5}$ and primary gaseous pollutant concentrations in Steubenville by severely inhibiting the vertical dispersion of pollutants. Conversely, summertime $PM_{2.5}$ episodes are more predominantly driven by the photochemical formation and transport of secondary $PM_{2.5}$. This hypothesis will be studied in greater depth using hourly air monitoring data and upper air observations in a future paper.[35]

Correlations among $PM_{2.5}$, Gases, and Weather Conditions

To further investigate and quantify the associations among $PM_{2.5}$, gaseous co-pollutants, and meteorological conditions that were observed above, we used simple linear regression analyses. As discussed, adjusted values were used in these analyses to ensure that autocorrelation and seasonal patterns had been removed from each data set being considered.

The second column of Table 3 presents the R^2 and slope values resulting from simple linear regression analyses of $PM_{2.5}$ as a function of each gaseous co-pollutant and meteorological condition measured at the Steubenville monitoring site, using all pairwise data collected during SCAMP. Regression model intercepts were also estimated but are not shown. The results indicate that even after removal of the autocorrelation, daily fluctuations in $PM_{2.5}$ concentrations at Steubenville were significantly associated (at the $\alpha = 0.05$ level) with daily fluctuations in the levels of each gaseous co-pollutant and weather condition studied, except for RH. Associations between $PM_{2.5}$ and SO_2, NO, NO_2, and CO were positive and relatively strong; adjusted values of each of these gaseous co-pollutants accounted for 25–46% of the variance in adjusted $PM_{2.5}$ values. Conversely,

associations between $PM_{2.5}$ and O_3, solar radiation, and BP were relatively weak, with R^2 values <0.05. Temperature exhibited a statistically significant positive association with $PM_{2.5}$, consistent with the results presented in Figure 2, and wind speed exhibited a statistically significant negative association with $PM_{2.5}$, consistent with the results shown in Figures 3 and 4.

When examining correlations among various air monitoring data, it is also important to consider the potential for lagged relationships. This is efficiently accomplished by use of cross-correlation function (CCF) plots. Two CCF plots are presented in Figure 6; these show Pearson correlations between adjusted BP values (Figure 6a) or adjusted temperature values (Figure 6b) and various lags of adjusted $PM_{2.5}$ values. CCF plots were prepared for all of the variable pairs considered in Table 3, and BP and temperature exhibited the strongest correlations with lagged $PM_{2.5}$. Figure 6a indicates that, in general, the BP on a given day at Steubenville tended to be positively associated with the $PM_{2.5}$ concentration experienced on the following day but negatively associated with the $PM_{2.5}$ concentration experienced on the previous day. This is consistent with the situations shown in Figures 3 through 5, in which an increase in BP precedes an increase in $PM_{2.5}$ concentration by ~1 day, and a decrease in $PM_{2.5}$ concentration with the passage of a frontal system is followed by an increase in BP ~1 day later. As indicated in Figure 6b, temperatures at Steubenville tended to be positively associated with the $PM_{2.5}$ concentrations experienced 1 day earlier. This is consistent with the behavior shown in Figure 5e, in which a rise and fall in $PM_{2.5}$ concentration is trailed slightly by a rise and fall in temperature. Hence, these CCF plots reveal and quantify the more complex nature of the associations between ambient levels of $PM_{2.5}$ and meteorological processes, which are missed if only correlations among paired daily data are studied. The results of CCF analysis are important for incorporation into models that attempt to predict ambient levels of $PM_{2.5}$ or quantify its health effects.

Although seasonal trends were removed from the adjusted datasets of each individual variable measured at Steubenville, the correlation between adjusted data for two distinct variables might still be seasonally dependent if the processes responsible for the observed association vary seasonally. We therefore examined the possibility of seasonal variations in the associations between daily average $PM_{2.5}$ and gaseous co-pollutants and weather conditions by repeating the simple linear regression analyses shown in the second column of Table 3, using subsets of adjusted data grouped by astronomical season. For each independent variable, the slopes of regression lines for spring, summer, and fall were compared with the slope of the regression line for winter by a statistical significance test. Winter was chosen as the base case for these comparisons because it is one of the two most climatologically extreme seasons and because correlations between independent variables and $PM_{2.5}$ tended to be more similar for winter, spring, and fall than for summer, spring, and fall. Hence, it was generally more interesting to test whether slopes in spring and fall were significantly different from slopes in winter than to test whether they were significantly different from slopes in summer.

The results presented in Table 3 show that the strengths of associations between $PM_{2.5}$ and several co-pollutants and weather conditions were substantially dependent on season, as R^2 values describing these associations varied by an order of magnitude or more from season to season in some instances. Additionally, statistically significant changes in the slope of the linear regression line with season were observed for several independent variables, indicating seasonal dependencies in the sensitivities of the associations between $PM_{2.5}$ and these variables. Graphical examples that clearly illustrate these seasonal variations in associations are provided in Figure 7.

As shown in Table 3, daily $PM_{2.5}$ concentrations measured during SCAMP were much more strongly and positively associated with daily concentrations of CO and especially NO and NO_2 during the fall and winter than during the summer. The adjusted values for each of these gases accounted for 53–61% of the variance in adjusted $PM_{2.5}$ values during the fall, but accounted for only 0.14–24% of the variance during the summer. This pattern is consistent with the abovementioned observation that episodic associations between $PM_{2.5}$ and primary co-pollutant gases were generally strongest during cooler periods of the year.

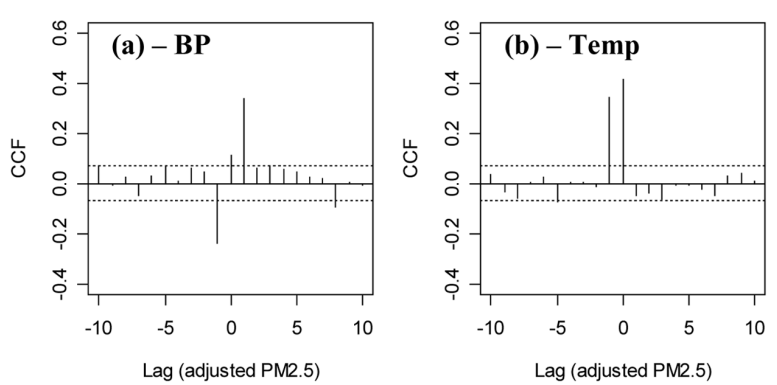

Figure 6. CCF plots showing Pearson correlations of adjusted BP values (a) and adjusted temperature values (b) with various lags of adjusted $PM_{2.5}$ values at Steubenville. Lags are in units of days. A negative lag of *n* days indicates that $PM_{2.5}$ occurs *n* days before the second variable being considered. Lines extending beyond the dotted interval indicate statistically significant correlations.

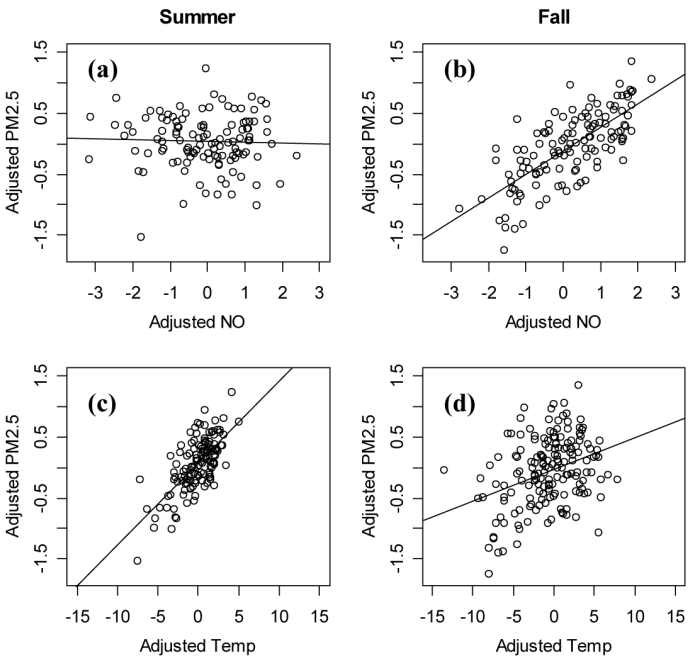

Figure 7. Examples of the dependence of strength of association on season. Scatter plots and linear regression lines are shown for adjusted $PM_{2.5}$ as a function of adjusted NO for summer (a) and fall (b), and for adjusted $PM_{2.5}$ as a function of adjusted temperature for summer (c) and fall (d).

Seasonal variations in the association between SO_2 and $PM_{2.5}$ were not as pronounced, with R^2 values ranging from 0.24 for the summer to 0.48 for the fall. Daily average temperatures were more strongly and positively associated with daily average $PM_{2.5}$ concentrations during the summer than during the other three seasons, whereas daily average wind speeds were more strongly and negatively associated with $PM_{2.5}$ during the fall, winter, and spring than during the summer. O_3 exhibited a relatively weak positive association with $PM_{2.5}$ during the summer and a relatively weak negative association during the other three seasons. The strong positive temperature association and positive O_3 association during summertime might reflect the relative importance of photochemically induced secondary $PM_{2.5}$ episodes during this season. However, the association between solar radiation and $PM_{2.5}$ was stronger and more positive during autumn than during summer.

The seasonal patterns discovered in associations among $PM_{2.5}$, gaseous co-pollutants, and weather conditions at Steubenville are similar to those reported by Chock et al.[26] among PM_{10} and various co-pollutant variables at Pittsburgh. As mentioned in the introduction, because of these seasonal patterns, Chock et al. were compelled to use seasonal models to study associations between pollutant concentrations and mortality, suggesting that even the typical filtering used in Poisson regression models likely fails to adequately remove seasonal dependence from all variables. They reported that although single and multipollutant nonseasonal models consistently indicated a significant association between PM_{10} and mortality, the use of seasonal models revealed collinearity problems among correlated concentrations of PM_{10}, CO, and NO_2, which cast substantial doubt on the non-seasonal modeling results. In light of these findings, strong seasonal patterns discovered in the correlations between $PM_{2.5}$ and CO, NO_2, and NO during SCAMP suggest that epidemiological studies assessing the health effects of PM in Steubenville should use seasonal models that carefully consider the effects of each of these pollutants. Past epidemiological studies that associated airborne PM with adverse health effects in Steubenville have not adequately done this. Schwartz and Dockery[23] and Moolgavkar et al.[24] included SO_2 in their analyses, and Moolgavkar et al.[24] applied seasonal models to examine associations among TSP, SO_2, temperature, and mortality at Steubenville; however, these analyses did not consider the effects of CO and NO_x, whose correlations with $PM_{2.5}$ exhibited a much stronger seasonal dependence than those of SO_2 during SCAMP. The Six Cities Study[9] considered SO_2, NO_2, and O_3 as predictors of mortality, but did not consider CO, which exhibited the strongest overall association with $PM_{2.5}$ of the co-pollutants measured during SCAMP, and which has been identified as a potentially significant predictor of mortality.[36] Moreover, due in large part to limitations related to the design of the Six Cities Study, neither the original analysis[9] nor a reanalysis[25] attempted to fit multipollutant models to the data to differentiate the effects of the various pollutants monitored. In their analysis of associations between fine and coarse particles and daily mortality in Steubenville, Schwartz et al.[37] failed to incorporate any gaseous pollutants, claiming that "adding in collinear gaseous pollutants would only confuse the paper and the analyses."

Hence, the seasonally dependent associations observed between $PM_{2.5}$ and co-pollutants during SCAMP illustrate the need to exercise caution when interpreting the results of past epidemiological studies at Steubenville that failed to thoroughly consider the potential confounding effects of co-pollutants and their possible seasonal correlations with PM. A future epidemiological study might attempt to better assess the health effects of the ambient air pollution mixture in Steubenville by incorporating the growing abundance of PM and gaseous co-pollutant data available for the city, including data collected as part of SCAMP.

Correlations among $PM_{2.5}$ Components, Gases, and Weather Conditions

To gain further insight into the associations observed between $PM_{2.5}$ and gaseous co-pollutant and weather variables at Steubenville, we examined associations between

Table 4. Simple linear regression results for adjusted $PM_{2.5}$ component values as a function of adjusted gaseous co-pollutant or meteorological condition values measured during SCAMP.

	SO_4^{2-}	NO_3^-	NH_4^+	EC	OM
SO_2					
R^{2a}	+0.22	+0.00	+0.10	+0.31	+0.22
p^b	**<0.0001**	0.8963	**0.0002**	**<0.0001**	**<0.0001**
NO					
R^2	+0.06	+0.07	+0.04	+0.38	+0.20
p	**0.0142**	**0.0049**	**0.0322**	**<0.0001**	**<0.0001**
NO_2					
R^2	+0.13	+0.06	+0.07	+0.39	+0.27
p	**0.0003**	**0.0161**	**0.0082**	**<0.0001**	**<0.0001**
CO					
R^2	+0.17	+0.04	+0.17	+0.44	+0.28
p	**<0.0001**	**0.0204**	**<0.0001**	**<0.0001**	**<0.0001**
O_3					
R^2	−0.00	−0.02	−0.00	−0.07	−0.01
p	0.7900	0.1780	0.6500	**0.0057**	0.3486
WS					
R^2	−0.02	−0.07	−0.01	−0.10	−0.14
p	0.1000	**0.0021**	0.2330	**0.0001**	**<0.0001**
Temp					
R^2	+0.27	−0.00	+0.15	+0.19	+0.28
p	**<0.0001**	0.5056	**<0.0001**	**<0.0001**	**<0.0001**
RH					
R^2	+0.01	+0.03	+0.02	−0.02	−0.03
p	0.173	**0.0272**	0.0984	0.1064	**0.0322**
Rad					
R^2	+0.00	−0.01	−0.00	+0.02	+0.03
p	0.9906	0.2853	0.6200	0.0907	**0.0439**
BP					
R^2	−0.01	−0.00	−0.01	+0.00	+0.00
p	0.2899	0.8511	0.3399	0.6999	0.8510

Notes: [a]The sign for R^2 refers to the slope of the regression line; [b]p value resulting from a test of the hypothesis that the slope of the regression line is equal to zero. Statistically significant p values are shown in bold; WS = wind speed; Temp = temperature; Rad = solar radiation.

particular $PM_{2.5}$ components and these variables. Table 4 shows simple linear regression results for adjusted values of various $PM_{2.5}$ components as a function of adjusted gaseous co-pollutant or meteorological condition values. Associations of O_3, RH, solar radiation, and BP with each of the $PM_{2.5}$ components were either statistically insignificant or relatively weak. SO_2, NO, NO_2, and CO were most strongly associated with the EC component of $PM_{2.5}$. Adjusted values of each of these gases were able to account for between 31% (SO_2) and 44% (CO) of the variance in adjusted EC values. EC aerosol originates from primary source emissions; hence, these findings possibly suggest the importance of common combustion sources in influencing ambient concentrations of these five chemical species in the Steubenville region. SO_2, NO, NO_2, and CO also exhibited rather uniform positive associations (R^2 = 0.20–0.28) with OM, suggesting that appreciable emissions of organic aerosol might originate from these same combustion sources. This is consistent with the finding of Connell et al.[22] that adjusted values of EC and OM measured during SCAMP were significantly and positively correlated (R^2 = 0.50).

SO_2 is a precursor to the secondary formation of SO_4^{2-} aerosol, and NO_x is a precursor to the secondary formation of NO_3^- aerosol. Although statistically significant positive associations were observed between adjusted values of these secondary aerosol species and their gaseous precursors, the associations were relatively weak, especially for NO, NO_2, and NO_3^-. This is likely a result of the influence of meteorological and other factors that affect the rates and equilibria of reactions governing secondary aerosol formation and to differences in the transport and deposition rates of the various gaseous and particulate species. Adjusted temperature values were most strongly associated with adjusted sulfate and OM values, perhaps reflecting the positive influence of temperature on the rates of the reactions leading to the secondary formation of these aerosol species. The statistically significant, positive association observed between EC and temperature cannot be interpreted in this fashion because EC is a primary aerosol species. Rather, this association might be indicative of temperature acting as a surrogate for atmospheric conditions that favor the accumulation of various pollutant species, as discussed earlier.

We also investigated the potential for seasonal variations in the correlations among fine particle components and gas and weather variables and discovered some statistically significant seasonal dependencies. For example, linear regression of adjusted OM as a function of adjusted NO yielded a slope of 0.11 and a R^2 of 0.33 for wintertime data. The slope and R^2 decreased to 0.0026 and 0.0002, respectively, when summertime data were used. Seasonal variations in correlations between particular $PM_{2.5}$ components and gas and weather variables did not provide conclusive explanations for the seasonal variations observed in the correlations between total $PM_{2.5}$ and these variables, perhaps in part because $PM_{2.5}$ composition was determined only every fourth day. Nonetheless, to more fully understand the nature and possible health effects of the ambient air pollutant mixture, future studies should focus more attention on associations among individual aerosol components, co-pollutants, and weather conditions.

Multiple Regression Time-Series Model for $PM_{2.5}$

Associations among individual gas or weather variables and $PM_{2.5}$ concentrations measured at Steubenville during SCAMP have been examined in depth. However,

many of the gas and weather variables are strongly associated with each other as well as with $PM_{2.5}$. It is possible therefore that a gas or weather parameter that is unrelated to $PM_{2.5}$ might exhibit a strong association with $PM_{2.5}$ in a simple linear regression model because both $PM_{2.5}$ and that parameter are related to a common third variable. To better understand the collective influence and relative importance of all of the gas and weather variables measured during SCAMP on $PM_{2.5}$ concentrations at Steubenville, we developed a single time-series model with external regressors to predict daily $PM_{2.5}$ concentrations as a function of past $PM_{2.5}$ concentrations and current and past gas concentrations and weather conditions.

Numerous models could have been formulated that would account for the variability in daily $PM_{2.5}$ concentrations to various extents. The model presented here, whose development was guided by the single-variable time-series models and simple linear regression results presented above, was the best (on the basis of R^2, Aikake Information Criterion, and satisfaction of modeling assumptions) of several candidate models. Log-transformed $PM_{2.5}$ concentrations at Steubenville were modeled by use of an order-one autoregressive model with multiple external regressors:

$$\ln(PM_{2.5}) = \omega_0 + (\omega_1 + \omega_2 S)\ln(NO_x) + \omega_3 \ln(CO + 0.1) + \omega_4 \ln(SO_2) + \omega_5 \sqrt{O_3} + (\omega_6 + \omega_7 S)Temp + \omega_8 \ln(WS) + \omega_9 RH^{1.5} + \frac{\varepsilon}{1 - \phi B} \quad (1)$$

where gas and weather variables are given by the symbols and in the units indicated in Table 2, ω_i and ϕ are parameters to be estimated, S is equal to 1 for summertime days and 0 for all other days, B is the backward shift operator, and ε is the random, normally distributed error. Note that the $1 - \phi B$ term is distributed across all model terms other than the error term, so that 1-day lags of each variable, including $PM_{2.5}$, are included in the model.

Estimates of all model parameters are shown in Table 5. The t statistics provided in Table 5 confirm that all of the parameters are statistically significant. Diagnostic plots of modeling results are presented in Figure 8. Model residuals were distributed rather normally (Figure 8b) and generally did not exhibit statistically significant autocorrelation (Figure 8c), especially at early lags. The randomness of the residuals is further confirmed by the Ljung–Box test results plotted in Figure 8d, which indicate no statistically significant associations among the first 10 lags. Log-transformed $PM_{2.5}$ concentrations predicted by the model are plotted against corresponding observed log-transformed $PM_{2.5}$ concentrations in Figure 8a. The points are closely and randomly scattered about the parity line, indicating that the model was successful in accounting for a substantial amount of the variability in $PM_{2.5}$ concentrations measured at Steubenville. The overall R^2 of the model was 0.79.

Table 5. Estimates and t statistics for parameters in eq 1.

Parameter	Estimate	t statistic
ω_0	1.08	4.12
ω_1	0.169	3.12
ω_2	−0.355	−8.28
ω_3	0.290	8.83
ω_4	0.210	7.20
ω_5	0.138	5.58
ω_6	0.0112	3.50
ω_7	0.0560	9.66
ω_8	−0.229	−4.14
ω_9	6.17×10^{-4}	6.00
ϕ	0.477	9.80

Rizzo et al.[7] developed similar time-series models with external regressors to predict decreases in ambient levels of PM_{10} that might result from corresponding decreases in O_3 because of reductions in O_3 precursor (i.e., NO_x and volatile organic compound) emissions. It is difficult to place confidence in such predictions, however, when it is uncertain whether a purely causal relationship exists between the variables being considered.

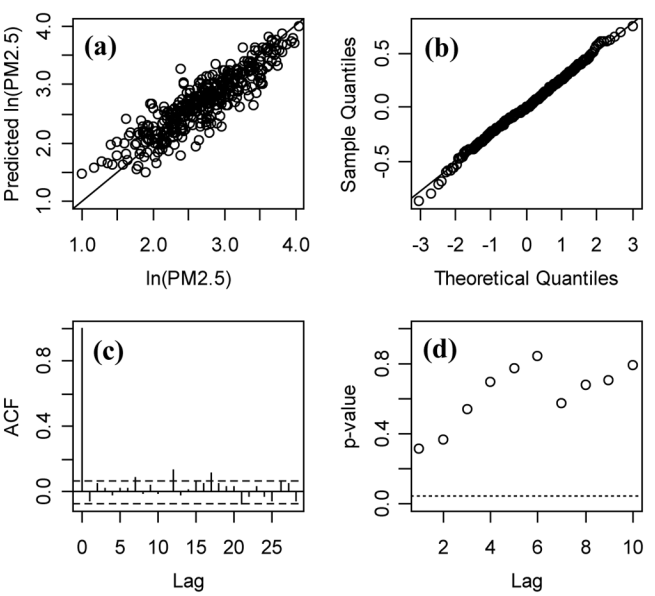

Figure 8. Results of the multiple regression time-series model for $\ln(PM_{2.5})$ as a function of transformed gaseous co-pollutant and weather data: (a) predicted vs. observed values with parity line, (b) normal probability plot of residuals, (c) ACF plot of residuals, (d) p values for the Ljung–Box statistic for various lags of residuals. For plots c and d, lags are in units of days. Lines extending beyond the dashed interval in plot c indicate statistically significant autocorrelations. The dotted line in plot d denotes $p = 0.05$.

For example, the positive association between O_3 and $PM_{2.5}$ in the model presented above might indicate the participation of O_3 or its precursors in photochemical reactions leading to secondary $PM_{2.5}$ formation, in which case a reduction in O_3 precursor emissions would likely result in a reduction in $PM_{2.5}$. Conversely, this association might merely reflect the influence of meteorological conditions on ambient levels of both $PM_{2.5}$ and O_3, in which case a reduction in O_3 precursor emissions would have no effect on $PM_{2.5}$ concentrations. The results in Figure 8a show that the model is a good predictor of daily average $PM_{2.5}$ concentrations at Steubenville. However, values of gaseous pollutant concentrations and weather conditions must be known for the day for which the $PM_{2.5}$ concentration is being predicted; hence, the model has limited use as a forecasting tool.

The model is useful, however, for studying the potential for confounding among pollutant and weather variables at Steubenville. Each statistically significant predictor variable is able to account for variability in $PM_{2.5}$ concentrations that could not be accounted for by any of the other variables in the model. For example, NO_x originates from many of the sources that emit either CO or SO_2. However, all three of these variables are statistically significant in the model, indicating that each is able to account for variability in $PM_{2.5}$ concentrations over and above that accounted for by the other two variables. Conversely, neither BP nor lagged BP is statistically significant in the model, suggesting that the association between BP and $PM_{2.5}$ can be fully accounted for by other variables, such as temperature and wind speed, that fluctuate with the passage of frontal systems. Seasonal patterns in correlations were again significant, although only for NO_x and temperature. Surprisingly, RH, which was not significantly associated with $PM_{2.5}$ in simple linear regression models, was a significant positive predictor in the multiple regression model.

The model presented above reveals that 79% of the variance in daily log-transformed $PM_{2.5}$ concentrations measured at Steubenville during SCAMP could be accounted for by other factors, including gaseous co-pollutant concentrations, weather conditions, and past $PM_{2.5}$ concentrations. This again emphasizes the need for epidemiological studies focusing on $PM_{2.5}$ in Steubenville (and elsewhere) to thoroughly consider the effects of many different gaseous co-pollutants and meteorological variables.

CONCLUSIONS

Daily average concentrations of $PM_{2.5}$, ionic and carbonaceous components of $PM_{2.5}$, and gaseous co-pollutants were determined at a monitoring site in Steubenville, OH, between May 2000 and May 2002. Daily average meteorological conditions and pollen and mold spore counts were also measured.

Analysis of these data suggests that meteorology is important in influencing ambient concentrations of $PM_{2.5}$ in Steubenville. Episodes of elevated $PM_{2.5}$ concentrations frequently consisted of an increase in concentration during a period of locally high pressure or elevated temperature, followed by a rapid decrease in concentration with the passage of a frontal system. The positive association with temperature was strongest during summertime. Wind speed often exhibited a negative association with $PM_{2.5}$ concentration, especially during cool portions of the year. The correspondence between high $PM_{2.5}$ concentrations, high BPs, low wind speeds, and the absence of frontal systems likely indicates the relationship between poor ventilation conditions and elevated $PM_{2.5}$ concentrations in Steubenville. Among the $PM_{2.5}$ components studied, temperature was most strongly associated with sulfate and OM, suggesting its possible effect on the rates of secondary formation of these species

Concentrations of CO, NO_x, and SO_2 were positively and significantly associated with concentrations of $PM_{2.5}$. Several $PM_{2.5}$ episodes during cool portions of the year were accompanied by elevated concentrations of all three of these gaseous co-pollutants, and associations between $PM_{2.5}$ concentrations and concentrations of CO and NO_x displayed a statistically significant dependence on season, with the strongest associations observed during fall and winter and the weakest associations observed during summer. The associations among $PM_{2.5}$, CO, NO_x, and SO_2 could result from common local source emissions of all four of these pollutants or from the presence of reduced mixing heights during cool-season inversions that concentrate $PM_{2.5}$ and primary gaseous pollutants near ground level. CO, NO_x, and SO_2 were most strongly correlated with the EC and OM components of $PM_{2.5}$, suggesting the possible importance of a common combustion source emission of these species. Ozone concentrations were positively associated with $PM_{2.5}$ concentrations during the summer and negatively associated with $PM_{2.5}$ concentrations during all other seasons, possibly reflecting the increased importance of photochemical $PM_{2.5}$ formation during the summer.

Although meteorological conditions appear to be important in affecting ambient $PM_{2.5}$ concentrations in Steubenville, meteorology obviously cannot be controlled to achieve reductions in $PM_{2.5}$. Rather, associations observed between $PM_{2.5}$ and meteorology indicate that if a $PM_{2.5}$ reduction strategy is required for Steubenville, the effects of weather conditions on $PM_{2.5}$ accumulation, transport, deposition, and secondary formation must be

carefully considered to assess whether a particular emission reduction will actually produce the anticipated decrease in ambient $PM_{2.5}$ levels. Seasonal patterns observed in associations between $PM_{2.5}$ concentrations and various gaseous co-pollutant concentrations and weather conditions suggest that mechanisms governing ambient levels of $PM_{2.5}$ and other pollutants might vary appreciably by season, such that different control strategies would be required for different times of year.

The relatively strong associations observed among daily $PM_{2.5}$ concentrations, gaseous co-pollutant concentrations, and weather conditions also highlight the potential for collinearity problems in epidemiological studies focusing on the health effects of $PM_{2.5}$ in Steubenville. NO_x, CO, SO_2, O_3, temperature, RH, and wind speed were all significant predictors of $PM_{2.5}$ concentration in a multiple regression time-series model, which was able to explain 79% of the variance in daily log-transformed $PM_{2.5}$ concentrations measured at Steubenville during SCAMP. Additionally, strong seasonal variations in the associations between $PM_{2.5}$ and CO, NO, and NO_2 illustrate the need for epidemiological studies to use seasonal models that consider all of these potentially confounding variables. These findings indicate that the results of previous PM epidemiology studies in Steubenville, which did not sufficiently consider the possible effects of co-pollutants such as CO, should be interpreted cautiously. Additionally, they suggest that a new epidemiological study should be conducted to more properly evaluate the health effects of the ambient air pollution mixture in Steubenville.

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